Reversible Transition between Perpendicular and In-Plane Magnetization in Ultrathin Films

D. P. Pappas, K.-P. Kämper, and H. Hopster
Department of Physics and Institute for Surface and Interface Science, University of California, Irvine, California 92717
(Received 11 April 1990)

The magnetization in ultrathin Fe layers (2.5–3.5 atomic layers) on Cu(100) reversibly switches between perpendicular (at low temperature) and in-plane magnetization (at higher temperature). The switching temperature decreases with increasing film thickness. The switching transition is attributed to the temperature dependence of the perpendicular anisotropy. The transition is accompanied by a loss of magnetization over a temperature range of 20–30 K and shows evidence for a canted-spin configuration.

PACS numbers: 75.70.Ak, 75.25.+z, 75.30.Gw

Magnetization in ultrathin films (1–10 atomic layers) has become a very active field of research recently.[1] These systems serve as models in the study of fundamental properties of magnetism in two dimensions. Since an isotropic 2D system at finite temperature has no long-range order,[2] two-dimensional ferromagnetism is dominated by magnetic anisotropies; e.g., a uniaxial anisotropy induces Ising-like behavior.[3] The existence of perpendicular uniaxial anisotropy is a natural consequence of the reduced symmetry at an interface.[4] If the perpendicular anisotropy is large enough, it can overcome the shape anisotropy and force the magnetization to be perpendicular to the plane of the film.[5] In addition, epitaxial growth of films on various substrates allows the stabilization of novel structural phases that are not realized in the bulk. These systems can be used to study the subtle interplay between structure and magnetism.[6] With the progress in sample-preparation techniques (molecular-beam epitaxy in ultrahigh vacuum) together with the development of surface-sensitive magnetometry and progress in electronic-structure calculations, there has been renewed interest in the search for well-defined epitaxial systems with large interfacial anisotropies.[7]

The magnetism of Fe films on Cu(100) and Ag(100) has been studied by many techniques over the last few years and, although much controversy exists on the details concerning the growth mode and quality of the films,[8] it seems to be well established now that these systems have a dominant perpendicular anisotropy at low temperatures.[9]

In this Letter we study the temperature-dependent magnetization of films a few atomic layers thick by spin-polarized secondary-electron spectroscopy. We find a transition ("switching") between perpendicular and in-plane magnetization. Under appropriate sample-preparation conditions the switching transition is perfectly reversible and is accompanied by a characteristic loss of magnetization over a temperature range of 20–30 K.

The experiments were performed in a new UHV system for spin-polarized secondary-electron spectroscopy. Low-energy secondary electrons (0–1 eV), excited by a 1-keV primary electron beam (0.5 mm diameter), are energy selected by a 90° spherical analyzer and their spin polarization is then measured in a medium-energy (30 keV) retarding-field Mott detector[10] using a uranium foil as a scattering target.[11] The spin polarimeter consists of two orthogonal detectors so that the full vector polarization can be measured. The electron beam is deflected electrostatically between the two detectors. In addition, the apparatus is equipped with a cylindrical-mirror analyzer for Auger-electron spectroscopy and a low-energy electron-diffraction (LEED) optics for structural characterization.

The films were evaporated from a chunk of high-purity Fe by electron bombardment. The evaporator is enclosed in a water-cooled jacket. The base pressure in the system is in the 10^{-11}-Torr range and during Fe evaporation the pressure remains below 3×10^{-10} Torr. Evaporation rates, monitored by a quartz microbalance, were typically 1–2 Å/min. The Cu crystal was oriented to within 1° by Laue diffraction, spark cut, and mechanically polished. It was cleaned by noble-gas-ion sputtering and annealing cycles until contamination levels were below the Auger detection limit ($<1\%$) and LEED showed a sharp 1×1 pattern.

Following the suggestions of Steigerwald, Jacob, and Egelhoff[12] the Fe films were deposited with the Cu sample at 100 K in order to minimize interdiffusion and agglomeration. This results in a very diffuse LEED pattern. The contamination of the films (mainly oxygen) is less than 3%. The samples were then annealed to 300 K whereupon the LEED spots sharpened and (5×1) superstructure spots appeared. This structure was then stable over the whole temperature range (100–350 K) in which the switching behavior was investigated. Annealing to above 350 K results in an irreversible transition to a sharp (2×1) reconstruction. Reconstructions have been reported in other studies,[13] albeit for different preparation conditions. Daum, Stuhlmann, and Ibach report that the reconstructions are suppressed by small amounts of oxygen contamination.[13] It is interesting to note that despite the diffuse LEED pattern at 100 K, indicating

poor epitaxy, the Auger uptake curve, i.e., the normalized iron signal, Fe(650 eV)/[Fe(650 eV)+Cu(850 eV)], as a function of evaporation time, shows distinct "breaks" which coincide with the breaks from films grown at 300 K. In accord with common practice, we assumed that the first break corresponds to the completion of one atomic layer (AL), and all thicknesses given are referenced to this calibration.

Before presenting the magnetic measurements it is important to state the exact experimental conditions. The Fe films are magnetized by current pulses through two orthogonal coils close to the crystal. One coil is oriented along the surface normal (z direction) for perpendicular magnetization and the other coil (x direction) supplies an in-plane magnetic field along a ⟨010⟩ crystal axis. During the measurements the sample is first magnetized in plane and the in-plane spin-polarization components (x and y) are measured. After a perpendicular magnetizing pulse the normal polarization (z direction) is measured. All polarization measurements are performed on the remanent magnetization, i.e., in zero external field.

In Fig. 1 the switching of the magnetization is demonstrated for 2.5- and 3.0-AL films. The magnetization curves clearly show four regions: At low temperature the films are magnetized strictly perpendicular (z direction), followed by a transition region where the magnetization is essentially zero. Above the transition the magnetization is mainly in plane (x direction) with an admixture of z polarization slightly above the transition. Heating to above 350 K results in an *irreversible* loss of magnetization. This is also the temperature where the LEED sharpens and changes irreversibly. We therefore attribute the loss of magnetization around 350 K to a structural phase transition, possibly into antiferromagnetic fcc Fe, which is the preferred growth mode at elevated temperatures.[14] This is corroborated by the fact that subsequent Fe deposition onto the film does not restore the magnetization. The Auger intensities do not change significantly upon annealing to 350 K and therefore we believe that effects of agglomeration and intermixing are small up to this temperature.

We stress, however, that the switching behavior over the temperature range of 100–350 K is perfectly *reversible*. The films could be cycled many times through the transition. The magnetic behavior did not change over several hours except for a slight decrease in the absolute polarizations ($< 5\%$) which can be attributed to contamination from the residual gas. We also note that the switching transition does not seem to be related to a structural transition because after the first anneal the (5×1) structure is stable, and no changes in the LEED can be detected by visual observation when cycling through the transition. We therefore attribute the switching transition to the intrinsic temperature dependence of the magnetic anisotropies in the film.

As seen from Fig. 1 the switching temperature T_s depends very sensitively on the thickness of the film. In Fig. 2 we show T_s for a collection of ten films. T_s is

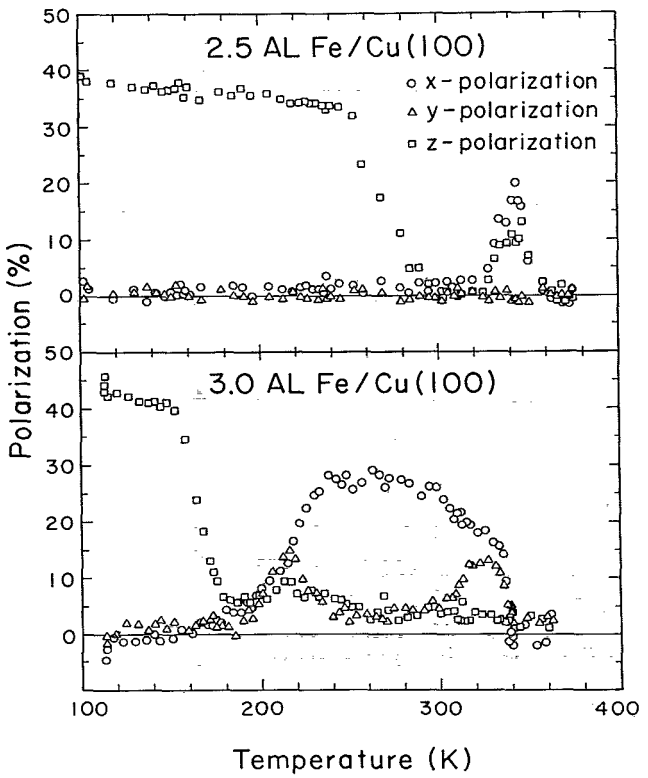

FIG. 1. Temperature dependence of the spin polarization of secondary electrons from two Fe films with slightly different thicknesses: 2.5 AL (upper panel); 3.0 AL (lower panel). See text for details.

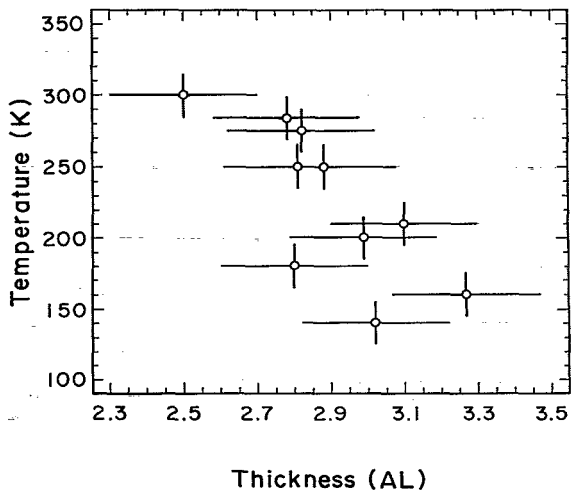

FIG. 2. Thickness dependence of the transition temperature T_s ("switching temperature") between perpendicular and in-plane magnetization.

defined as the center of the transition region. T_s shows a clear decreasing trend with increasing film thickness. This can be used to estimate the temperature dependence of the perpendicular anisotropy. If the switching temperature corresponds to the point where the perpendicular anisotropy K_u is compensated by the demagnetizing field, which is proportional to the thickness, then the anisotropy K_u decreases by about 20% between 100 and 300 K. This moderate temperature dependence is in qualitative agreement with recent calculations[15] and is, of course, the reason why the switching behavior can be observed only over a narrow thickness range.

The most striking and interesting feature in Fig. 1 is the complete loss of magnetization in the transition region. There are basically two different explanations for this behavior. First, at the temperature where the perpendicular and the shape anisotropy cancel the system essentially becomes isotropic and, therefore loses long-range order, i.e., is in a paramagnetic state. Above the transition long-range order emerges again (dominated by the shape anisotropy and in-plane anisotropies). The transition extends over a finite temperature range because at a *finite* temperature a minimum anisotropy is required to sustain long-range order.[4] In contrast to this dynamic picture, one can envision a static domain model where the loss of magnetization is due to a temperature-dependent domain size, the domains becoming microscopic over a certain temperature range. Yafet and Gyorgy[16] recently showed that in a ferromagnetic monolayer with perpendicular magnetization stripe domains are energetically favorable over a uniform magnetization. They showed that the width of the domains depends sensitively on the anisotropy and changes from microscopic to macroscopic over a narrow range.

These two pictures, dynamic spin fluctuations versus static domains, are not mutually exclusive, however. The system might actually go through both phases. Also, the domain picture becomes questionable when the domain size becomes comparable to the domain-wall width. It would be highly interesting to use secondary-electron microscopy with polarization analysis to spatially resolve any magnetic domain pattern and follow its evolution through the transition region.

As mentioned before, above the transition temperature, the polarization is not purely in plane. Close to the transition there exists a significant z-polarization component. We attribute this to a canted-spin configuration; i.e., the switching is a continuous turning of the magnetization direction from perpendicular to in-plane orientation. We also should comment on the occurrence of a significant y polarization (see lower panel of Fig. 1) in the temperature range above the transition and also between 300 and 340 K. These y polarizations were typically observed in these films. We believe that they are indicative of temperature-dependent in-plane anisotropies and possibly a tendency towards domain formation in the in-plane–magnetized films. This would also explain that the observed polarizations for the in-plane films are consistently somewhat smaller than the z polarizations.

In summary, we have shown that the temperature dependence of the perpendicular magnetic anisotropy leads to a reversible transition between normal and in-plane magnetization in films of 2.5–3.5 atomic layers of Fe on Cu(100). The transition is accompanied by a loss of macroscopic remanent magnetization. This is attributed to the loss of long-range order in an isotropic system and/or the temperature dependence of the magnetic domain size.

This work was supported by the National Science Foundation through Grant No. DMR 8821293 and by IBM Almaden Research Center.

[1]For a recent collection of review articles on the field, see Appl. Phys. A **49**, 1989 (special issue, edited by D. Pescia).

[2]N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).

[3]M. Bander and D. L. Mills, Phys. Rev. B **38**, 12015 (1988).

[4]L. Neél, J. Phys. Radium **15**, 376 (1954).

[5]U. Gradmann and J. Müller, Phys. Status Solidi **27**, 313 (1968).

[6]J. Kübler, Phys. Lett. **81A**, 81 (1981); C. S. Wang, B. M. Klein, and H. Krakauer, Phys. Rev. Lett. **54**, 1852 (1985); V. L. Moruzzi, P. M. Marcus, K. Schwarz, and P. Mohn, Phys. Rev. B **34**, 1784 (1986).

[7]B. T. Jonker, K.-H. Walker, E. Kisker, G. A. Prinz, and C. Carbone, Phys. Rev. Lett. **57**, 142 (1986); J. G. Gay and R. Richter, Phys. Rev. Lett. **56**, 2728 (1986).

[8]D. A. Steigerwald and W. F. Egelhoff, Jr., Phys. Rev. Lett. **60**, 2558 (1988); D. Pescia, M. Stampanoni, G. L. Bona, A. Vaterlaus, F. Meier, G. Jennings, and R. F. Willis, Phys. Rev. Lett. **60**, 2559 (1988); S. H. Lu, J. Quinn, D. Tian, F. Jona, and P. M. Marcus, Surf. Sci. **209**, 364 (1989).

[9]D. Pescia, M. Stampanoni, G. L. Bona, A. Vaterlaus, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 2126 (1987); B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, Phys. Rev. Lett. **59**, 1756 (1987); N. C. Koon, B. T. Jonker, F. A. Volkening, J. J. Krebs, and G. A. Prinz, Phys. Rev. Lett. **59**, 2463 (1987); M. Stampanoni, A. Vaterlaus, M. Aeschlimann, and F. Meier, Phys. Rev. Lett. **59**, 2483 (1987); J. R. Dutcher, B. Heinrich, J. F. Cochran, D. A. Steigerwald, and W. F. Egelhoff, Jr., J. Appl. Phys. **63**, 3464 (1988); J. J. Krebs, B. T. Jonker, and G. A. Prinz, J. Appl. Phys. **63**, 3467 (1988); C. Liu, E. R. Moog, and S. D. Bader, Phys. Rev. Lett. **60**, 2422 (1988).

[10]F.-C. Tang, X. Zhang, F. B. Dunning, and G. K. Walters, Rev. Sci. Instrum. **59**, 504 (1989).

[11]D. P. Pappas and H. Hopster, Rev. Sci. Instrum. **60**, 3068 (1989).

[12]D. A. Steigerwald, I. Jacob, and W. F. Egelhoff, Jr., Surf. Sci. **202**, 472 (1988).

[13]W. Daum, C. Stuhlmann, and H. Ibach, Phys. Rev. Lett. **60**, 2741 (1988); C. Egawa, E. M. McCash, and R. F. Willis,

Surf. Sci. **215**, L271 (1989).

[14]W. Keune, R. Halbauer, U. Gonser, and L. D. Williamson, J. Appl. Phys. **48**, 2976 (1977); W. Becker, H.-D. Pfannes, and W. Keune, J. Magn. Magn. Mater. **35**, 53 (1988); R. Halbauer and U. Gonser, J. Magn. Magn. Mater. **35**, 55 (1988); A. Amiri Hezaveh, G. Jennings, D. Pescia, R. F. Willis, K. Prince, M. Surman, and A. Bradshaw, Solid State Commun. **57**, 329 (1986); W. A. A. Macedo and W. Keune, Phys. Rev. Lett. **61**, 475 (1988).

[15]R. Richter and J. G. Gay, Mater. Res. Soc. Symp. Proc. **151**, 3 (1989).

[16]Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).